

Remarks

Claims 1-4, 7-14, 16, 18-27, 29, 31-38, 40-42, 44-46, 48-49, 51, 53, 55-58, and 60 were pending in the present application. Claims 1, 12, 24, 29, 36, 44, and 53 have been amended, and new claims 62-64 have been added. As a result of this amendment, claims 1-4, 7-14, 16, 18-27, 29, 31-38, 40-42, 44-46, 48-49, 51, 53, 55-58, 60, and 62-64 remain pending. Reexamination and reconsideration are requested in light of the accompanying remarks.

The objection to claim 29 has been overcome. Claim 29 has been amended to depend from claim 24.

The rejection of claims 1-4, 7-14, 16, 18-27, 29, 31-38, 40-42, 44-46, 48-49, 51, 53, 55-58, and 60 under 35 U.S.C. § 103(a) as obvious over, Nunan (U.S. Patent No. 6,040,265) has been overcome.

Nunan teaches a method of making a promoted support for improved catalysts for conversion of exhaust from internal combustion engines. The support comprises a mixed metal oxide promoter containing at least Ce and Zr substantially uniformly dispersed as homogeneous crystallites of less than about 100 Å on a high surface area refractory oxide support substrate. The method includes dissolving a combination of Ce- and Zr-oxide precursors, and optionally another metal oxide precursor, and a compatible organic depositing agent, slowly heating to transform the depositing agent into a gel-like matrix coating the substrate in which the mixed metal oxide precursor compounds are uniformly distributed and thereafter calcining to burn off the organic matrix and form the appropriate oxide morphology. Abstract.

According to the examiner, "Nunan teaches an apparatus for reducing an amount of carbon monoxide in process gas wherein the catalyst is a ceria based catalyst which promotes water gas shift reactions (col. 1, lines 43-56), the catalyst system comprising a noble metal/(Pt or Pd) (col. 10, line 65); a mixed metal oxide support consisting essentially of cerium oxide and zirconium oxide, wherein cerium oxide is present in an amount from about 45% to about 90% by weight of mixed metal oxide and zirconium/lanthanum oxide is present in an amount from about 10% to 55% by weight of mixed metal oxide (col. 10, lines [sic] 64 col. 11, lines 1-5); and passing the process fuel gas through the water gas shift converter in effective contact with the high activity water gas shift catalyst system and converting at least 50% of the carbon monoxide in the process fuel gas into carbon dioxide and hydrogen by a water gas shift reaction over a temperature range of about 300°C to about 450°C/(400°C to about 575°C) (see Figure 15).

Nunan does teach that his catalyst is for reducing an amount of carbon monoxide in process gas wherein the catalyst is a ceria based catalyst which promotes water gas shift reactions (col. 1, lines 43-56). Therefore it would be obvious to one having ordinary skill in the art to try placing Nunan's catalyst system in a "water gas shift reactor," or any "reactor", and pass there through a process gas stream as opposed to an exhaust stream for the purpose of achieving an entirely expected result - which is reducing carbon monoxide. . . . Furthermore, Nunan's catalyst is the same as that instantly claimed and therefore will perform as such."

Contrary to the examiner's statement, Nunan does not teach or suggest use of their catalyst in a water gas shift converter. Nunan teaches three way catalysts (TWC) for vehicle exhaust conversion. There are three reactions involved, including: 1) reducing nitrogen oxides to nitrogen and oxygen,  $2\text{NO}_x \rightarrow x\text{O}_2 + \text{N}_2$ ; 2) oxidation of carbon monoxide to carbon dioxide :  $2\text{CO} + \text{O}_2 \rightarrow 2\text{CO}_2$ ; and 3) oxidation of unburned carcinogenic hydrocarbons (HC) to carbon dioxide and water:  $2\text{C}_x\text{H}_y + (2x+y/2)\text{O}_2 \rightarrow 2x\text{CO}_2 + y\text{H}_2\text{O}$ . See col. 14, lines 27-39; col. 21, lines 17-60, and Tables 3-5, 7-8. These reactions are simultaneous when the A/F ratio is properly adjusted in order to have the proper amount of hydrocarbons to be converted on account of the  $\text{NO}_x$  which is being reduced.

These reactions are different from the water gas shift reaction, in which the reactants are carbon monoxide and water. The water gas shift reaction involves conversion of carbon monoxide and water to carbon dioxide and water:  $\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2$ . Paragraph [0007]. Thus, Nunan's patent, which involves different reactions taking place with different inlet gases and producing different outlet gases, does not teach or suggest using its motor vehicle exhaust conversion catalyst for water gas shift reactions.

The examiner's citation of col. 1, lines 43-56, does not indicate that Nunan's catalyst could be used in a water gas shift converter. This section primarily discusses prior art motor vehicle exhaust conversion catalysts. In addition, it simply states that it has been suggested that cerium might promote the water gas shift reaction.

*Motor vehicle exhaust conversion catalysts* are normally operated under conditions which inherently swing between oxidizing and reducing as an oxygen sensor and its control system keep the air/fuel ratio within the desired operating A/F window around the stoichiometric value. Ceria is a well-known component of *such exhaust conversion catalysts*. It is often referred to as an "oxygen storage" agent because it is

considered to have the ability to give up oxygen when the catalyst is exposed to reducing conditions and to re-oxidize when exposed to oxidizing conditions. It has also been suggested that ceria may stabilize the support structure, promote the activity of the precious metals, or promote the water gas shift reaction. See for example, B. Harrison, A. F. Diwell and C. Hallet, *Platinum Metals Rev.*, 1988, 32(2), 73-78.

Col. 1, lines 43-56. This is not a teaching or suggestion that Nunan's catalyst (or any other motor vehicle exhaust conversion catalysts) could be used in water gas shift converters.

Nunan's catalyst is an exhaust gas catalyst for *internal combustion engines*.

A method of making a unique, promoted support for improved catalysts for *conversion of exhaust from internal combustion engines*, said support comprising a mixed-metal-oxide promoter containing at least Ce and Zr substantially uniformly dispersed as homogeneous crystallites of less than about 100Å on a high-surface-area refractory oxide support substrate, comprising dissolving a combination of Ce- and Zr-oxide precursors, and optionally another metal oxide precursor, and a compatible organic depositing agent, slowly heating to transform the depositing agent into a gel-like matrix coating the substrate in which the mixed-metal-oxide precursor compounds are uniformly distributed and thereafter calcining to burn off the organic matrix and form the appropriate oxide morphology.

Abstract.

The present invention relates to catalysts for *conversion of exhaust from internal combustion engines*. More particularly, the present invention relates to mixed-metal oxide crystallite compositions supported and dispersed on a high surface area support such as  $\gamma\text{Al}_2\text{O}_3$ , their use as promoters in catalysts for motor vehicle exhaust conversion, and promoter catalyst supports containing mixed-metal oxide crystallite compositions.

Col. 1, lines 14-21.

It is an object of the present invention to provide an *exhaust conversion catalyst support material* that is highly effective in enhancing the performance of catalytically active metals deposited thereon in *the conversion of noxious components in the exhaust from internal combustion engines* to compounds that are not detrimental to the environment.

Col. 2, lines 46-51.

The resulting supported catalytic system is dried and calcined to form a *promoted exhaust conversion catalyst*. The *exhaust conversion catalyst* can then be washcoated onto a suitable carrier to form a structure for use in catalytic converters.

Col. 4, lines 34-38.

Interestingly, the improvements are not generally observed when the catalyst has been freshly prepared, but become apparent generally only after the catalyst has been exposed to reducing conditions as occur during normal usage in *automobile exhaust converters*.

Col. 7, lines 21-25.

Such homogeneous, small crystallite-sized, mixed-metal-oxide phases have unique and desirable applications in *exhaust conversion catalysts*.

Col. 9, lines 25-27.

*Exhaust conversion catalysts* made with the promoted supports of the present invention, when analyzed, show exceptionally small, homogeneous crystallites of the mixed-metal oxide essentially uniformly dispersed on the catalyst support. The properties of the mixed-metal-oxide crystallites of this invention were determined by X-ray diffraction (XRD), transmission electron spectroscopy (TEM), and temperature programmed reduction (TPR) analysis. The analysis results generally showed that catalyst supports prepared by the methods of this invention have very thin outer layers of uniformly and well dispersed homogeneous crystallites, generally of a size averaging 100 Å or less, more often 50 Å or less and even as small as 35 Å or less, as measured by X-Ray diffraction (XRD) and transmission electron spectroscopy (TEM). The very small size, homogeneous crystalline nature and uniform dispersion of the mixed-metal-oxide-promoted supports of this invention provide superior catalyst performance.

Col. 9, line 56 to col. 10, line 6.

Engine evaluation of Pt,Rh catalysts prepared using this material was subsequently carried out as described in detail in Examples 13, 14 and 15 of U.S. Pat. No. 5,064,803. Besides the stand dynamometer evaluations, FTP test evaluations on a 2.5 L TBI Buick Somerset and a 3.1 L MPFI Cavalier were carried out. The experimental catalysts were compared to two Pt,Rh reference catalysts. The results of these comparisons are summarized in Tables 3-5 and FIGS. 21 to 23.

In Tables 3 and 4 are summarized the integral performance numbers and aged light-off activity for the catalyst of the present invention compared to a preparation having double the Ce loading and to a case where no solid solution was formed between the  $\text{CeO}_2$  and  $\text{ZrO}_2$  components but where the Ce and Zr loading were the same as for the test catalyst. Clear performance advantages are observed for the catalyst of the present invention. In Table 5 are summarized the FTP vehicle results and again we see advantages for the catalyst of the present invention.

Col. 25, lines 3-22. See also col. 26, lines 20-55; col. 28, lines 35-45; col. 29, lines 10-18; col. 32, lines 38-53; claims 1-2 ("method of preparing a promoted catalyst support effective in enhancing the performance of catalytically active metals in the *conversion of exhaust gas from internal combustion engines*. . . ."; and Figs. 1-2, and 16-25 which show hydrocarbon conversion as a function of the air to fuel (A/F) ratio.

The examiner stated that "[t]he instant claims fail to positively recite the method such that carbon dioxide and water are introduced into the reactor. . . . The fact that the instant claim reads on a method of reducing an amount of carbon monoxide in process fuel gas in a water gas shift converter, the claim language does not preclude treating exhaust gas as in Nunan since exhaust gas can be considered "a process fuel gas", for example in a burner."

Claims 1, 12, and 24 have been amended to recite a "method of reducing an amount of carbon monoxide in a feed stream for a fuel cell" including "introducing a hydrocarbon feed stream into a primary reactor and reacting the hydrocarbon feed stream in effective contact with a reforming catalyst forming primary reactor products containing hydrogen, carbon monoxide, carbon dioxide, and methane," "introducing the primary reactor products into the water gas shift converter in effective contact with the high activity water gas shift catalyst system, and reacting the carbon monoxide and water to form carbon dioxide and hydrogen using a water gas shift reaction forming the feed stream for the fuel cell; and introducing the feed stream into the fuel cell." Thus, method claims 1, 12, and 24 (and those that depend from them) preclude the treatment of exhaust gas from combustion engines, and the various reactions associated with it which are treated with TWC.

According to the examiner, "it would be obvious to one having ordinary skill in the art to try placing Nunan's catalyst system in a "water gas shift reactor" or any "reactor" and pass there through a process gas stream as opposed to an exhaust stream for the purpose of achieving an

entirely expected result - which is reducing carbon monoxide." However, obviousness requires a reasonable expectation of success (MPEP 2143.02), which is completely lacking in this case. Catalysts are developed for the specific application and reactions involved. One of skill in the art would not expect that a three way catalyst for treating exhaust gas from internal combustion engines would provide the claimed conversion rate in a water gas shift reaction. As discussed above, the gases and the reactions involved are different.

In addition, contrary to the examiner's statement, Nunan does not teach or suggest converting at least 50% of the carbon monoxide in the process fuel gas into carbon dioxide and hydrogen by a water gas shift reaction over a temperature range of about 300°C to about 450°C/(400°C to about 575°C) (see Figure 15)." Using the line shown in Fig. 15, the Ce,Zr support does not reach 50% conversion of CO until at least 350°C. The first data point showing 50% conversion for the Ce,Zr support is at 450°C. Note that there are no data points between 300°C and 450°C for the Ce/Zr support like are shown in the other graphs. It is unclear why the lines are shown the way they are in the absence of actual data. Moreover, neither the graph nor the corresponding discussion indicate that a "promoter comprising alkali metals, or alkaline earth metals, or combinations thereof" was present with any of the supports shown in Fig. 15. Thus, Fig. 15 does not teach or suggest the claimed conversion rates for the claimed compositions.

The examiner admitted that "the first data point showing 50% conversion for the Ce,Zr support is at 450°C and that there are no data points between 300°C and 450°C for the Ce/Zr support. . . ." However, the examiner stated that "the temperature of 450°C is touching the boundary of the instantly claimed range of about 300°C to about 450°C. Therefore, Nunan does teach converting at least 50% of the carbon monoxide in the process fuel gas into carbon dioxide and hydrogen by a water gas shift reaction over a temperature range of about 300°C to about 450°C."

First, Nunan's graphs of % conversion relate to its three way catalyst treating exhaust from combustion engines. Even assuming that Nunan teaches that its catalyst could be used for water gas shift reactions in col. 1, lines 43-57 (which Applicants deny for the reasons given above), Nunan's entire discussion and all of the results presented relate to the *three way catalyst that it describes and uses to treat the exhaust from combustion engines*. Nunan does not include any discussion or results involving the water gas shift reaction. All of the graphs showing % conversion include CO, HC, and NO (the three components being treated with the three way

catalyst in exhaust gas from combustion engines), which clearly demonstrates that the exhaust from combustion engines is being treated and measured. Thus, the % conversion shown in Fig. 15 is the *conversion due to the oxidation of carbon monoxide to carbon dioxide in the treatment of the exhaust from combustion engines using a three way catalyst*, not the conversion of carbon monoxide and water to carbon dioxide and water in the water gas shift reaction. Fig. 15 has nothing to do with the water gas shift reaction.

Moreover, claims 1, and 12 require that the "high activity water gas shift catalyst system converts at least 50% of the carbon monoxide in the primary reactor products into carbon dioxide and hydrogen by the water gas shift reaction over a temperature range from about 300°C to about 450°C," not just at one point. Nunan does not show any data on conversion between 300°C and 450°C.

The examiner further stated that "Nunan's disclosure teaches the presence of the promoter, as set forth in the rejection above, which applies to the examples discussed therein. . . . Therefore, the catalysts disclosed in Nunan, including the example of Figure 15, do comprise a promoter as set forth herein and above." However, the fact that promoters *can* be included in Nunan's catalyst does not mean that they *were* included in the catalysts tested in Fig. 15. The only composition information Nunan provides for the catalysts of Fig. 15 does not indicate the presence of any promoters. Therefore, it cannot be assumed that the results shown in Fig. 15 are for catalysts including a promoter.

Furthermore, the promoter in Nunan's catalyst is present in an amount of about 1-20 wt%. However, composition claims 36, 44, and 53 are limited to about 0.1% to about 0.2%, about 0.1% to about 0.2%, and about 0.1% to about 0.5% promoter, respectively. Support for the amendment to claim 36 can be found in Fig. 4, lines for 0.2% Cs and 0.2% Ir, Fig. 5, lines for 0.2%Cs/2%Pt and 2%Pt/0.2%Cs, Fig. 6, lines for 0.2% Cs, Fig. 7 all lines, and Fig. 8, line for 0.2% Cs. Support for the amendment to claim 44 can be found in Fig. 5, line for 0.2%Cs a9.6% CeO<sub>2</sub>/80.4%ZrO<sub>2</sub>. Support for the amendment to claim 53 can be found in Fig. 2, line for (1%Pt/0.5%Cs)/CeO<sub>2</sub>-La<sub>2</sub>O<sub>3</sub> (75%:25%), Fig. 3, line for (1%Pt/0.5%Cs)/CeO<sub>2</sub>-La<sub>2</sub>O<sub>3</sub> (25%:75%), and Fig. 6, line for (0.2%Cs/2%Pt)/La<sub>2</sub>O<sub>3</sub>-CeO<sub>2</sub> (8.6%:91.4%). Therefore, Nunan does not teach or suggest the claimed catalyst.

Consequently, there is no reasonable expectation that a three way catalyst having a specific composition developed for treating the exhaust gas of internal combustion engines would provide the claimed conversion rate in a water gas shift reaction.

Therefore, claims 1-4, 7-14, 16, 18-27, 29, 31-38, 40-42, 44-46, 48-49, 51, 53, 55-58, and 60 would not have been obvious to one of ordinary skill in the art at the time the invention was made over Nunan.

### CONCLUSION

Applicants respectfully submit that, in view of the above remarks, the application is now in condition for allowance. Applicant respectfully requests that the claims be passed to allowance.

If the Examiner has any questions or comments regarding the present application, he is invited to contact the undersigned attorney at the telephone number indicated below.

Respectfully submitted,  
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